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ARTICLE

The Flame Emission of Indium from a Pyrotechnical View

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Dedicated to Professor Hans-Jörg Deiseroth on the Occasion of his 75th Birthday

Abstract. Until today, all blue-colored light-generating pyrotechnics are still based on copper and a halogen-source providing the blue-emitting species copper(I) chloride, copper(I) bromide or copper(I) iodide. The use of indium as a potential halogen-free blue light emitter in modern pyrotechnics is described. Therefore, metallic indium was introduced as both fuel and colorant in various pyrotechnical formulations including guanidine nitrate or potassium nitrate as oxidizing agent as well as magnesium, hexamethylentetramine and 5-amino-1*H*-tetrazole as fuel. The effect of incandescence was examined by applying different magnesium contents within the mixtures. Emission spectra and occurring emission lines of indium-based pyrotechnical compositions were recorded and evaluated for the first time. Since the expected blue flame color could not be completely achieved, the emission of indium was discussed from an academic point of view.

Introduction

Since the earliest advances in pyrotechnical research, the most challenging task is the production of a saturated blue flame color.[1] Until now, there is no sufficient alternative material to copper and copper-containing chemicals.[2] In a typical blue flare formulation, these compounds were mixed with potassium perchlorate $KClO_4$ as oxidizing agent, some fuel and additional poly(vinyl chloride) (PVC) to generate the metastable copper(I) chloride as blue light emitter.[1,3] Blue colorants are for example Paris Green $Cu(CH_3COO)_2{\cdot}3Cu(AsO_2)_2$ or Scheele's Green $CuHAsO_3$. These components are high-performing and efficient, nevertheless, there are also high toxic according to their arsenic content.[4] Further, water soluble copper salts like copper nitrates suffer from aqueous toxicity and cause groundwater contamination.[5] The perchlorate anion is highly persistent in water, therefore it contaminates groundwater and soil each time it is used in fireworks or military approaches.[6] Because of its similar ionic radius as iodide, perchlorate inhibits the iodide uptake in the thyroid glade.[7] For this reason, the Environmental Protection Agency (EPA) added perchlorates to the Contaminant Candidate List, since this anion has been detected in both drinking water and groundwater affecting human's health.[8] Another problematic issue is the formation of toxic chlorinated organic compounds, such as polychlorinated dibenzo-*p*-dioxins (PCDD) and dibenzofurans (PCDF) during the combustion of commonly used organic binders and a chlorine source.[9]

To overcome these issues, *Koch* evaluated other copper(I) halides CuX (X = F, Br, I) by UV/Vis spectroscopy as alternative blue-emitting species as it has been suggested by *Douda*.[2a,10] At the same time, *Klapötke* et al. reported on chlorine-free blue flare formulations based on copper(II) iodate.[11] The mixture with copper(I) iodide as blue light emitter provides a spectral purity (SP) of 64 % and dominant wavelength (DW) of 477 nm. More recent results apply a mixture of copper(I) bromide and potassium bromate for generating a blue color impression.[12] Unfortunately, these formulations suffer from impact and friction sensitivity and the resulting analogous polybrominated organic combustion products are also believed to be toxic.[13] There is insufficient information about the environmental and health impact of polyiodinated organic compounds given in the literature, however, it is expected to be similar to other polyhalogenated materials.[14] Regarding blue strobes, there is even less information given in the literature.[15] A constant burning flame with high color purity and intensity is called flare, whereas compositions that show an oscillatory combustion are referred as strobes.[15a,16] In 2017, the established Jennings-White blue strobe system consisting of 55 % ammonium perchlorate, 30 % tetramethylammonium nitrate (TMAN) and 15 % copper was modified with various ingredients and additives to study the effect on the resulting strobing behavior.[15d]

Further research is mandatory to surpass the performance of the best performing flare formulations until now (DW = 465 ± 20 nm and SP ≥ 65 %) and to meet the requirements for much environmentally benign compositions. The claim for so-called "next generation pyrotechnics that reduce the environmental impact" was also emphasized by a Statement of Need of the Strategic Environmental Research and Development Program (SERDP) in 2017.[17] Especially perchlorate oxidizers should be banned in modern formulations. An excessive literature research revealed two other postulated ways to generate blue light without the need of halogen and copper atoms. Some literature mentioned cesium salts to give a pale blue color, which unfortunately does not meet the requirement for blue

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illumination signals in terms of DW and SP.[2a,18] The second hypothesis mentioned by Jennings-White is based on the observations published by *Reich* and *Richter* in 1863 about their discovery of indium.[19] They reported a violet color with two emission lines in the Bunsen burner and a blue light upon the addition of hydrochloric acid.

Indium is a silvery-white rare heavy metal. Its crustal abundance is similar to silver and mercury.[20] Unfortunately, it is produced exclusively as a by-product during the processing of tin and lead.[21] Due to the excessively high demand and at the same time low supply of indium, it is one of the scarcities raw materials on earth.[20b,21a,22] Obviously, indium would never be considered as a pyrotechnic colorant for mass production until now.[23] Today′s application of indium compounds is dominated by indium tin oxide (ITO) as an essential part of touch screen panels, flat screens and transparent layers in perovskite solar cells.[24] Next to *Reich* and *Richter* in 1863, the flame chemistry of indium was further studied by *Paschen* and *Campbell* later on, who detected additional emission lines and assigned them to the corresponding excitations.[25] *Reich* and *Richter* reported a blue light emission upon addition of hydrochloric acid. Due to the previously discussed health concerns, no chlorine source was considered for application in newly designed pyrotechnical formulations. Analogous to the copper(I) salts CuCl, CuBr and CuI, the corresponding indium(I) bromide or indium(I) iodide could be considered as potential colorants at first. Already in 1991, *Singh* et al. reported the main emission of InBr and InI to be in the green region (520 nm), which prohibits the application as blue colorant.[26] Consequently, it was concluded that it is not possible to generate deep blue light through the application of other halogen atoms. The high price and low availability of pure indium metal presumably explains why no further research was done on indium as a blue-light emitter in pyrotechnic formulations so far. Due to the known disadvantages of indium, the presented study to the emission of indium is mainly of academically interest.

Results and Discussion

Nevertheless, the idea was to investigate, if further tuning of the flame conditions in halogen-free indium-based formulations might shift the emission lines into the desired blue range. First experiments (Table 1) applied guanidine nitrate (GN) as a metal-free oxidizer (T_{dec}: ≥ 250 °C).[27] It's a favored oxidizing agent in gas- and smoke-generating pyrotechnical systems.[3b] Further, in 2015 *Sabatini* et al. discovered nitrogen-rich fuels like hexamethylentetramine (hexamine) in red-colored halogen-free flares to have a deoxidizing effect on the flame entailing higher luminosity and spectral purity.[28] Indium served as both colorant and fuel. The two-component epoxy binder system was a mixture of Epon 813 and Versamid 140 (ratio 4:1).

The amount of oxidizer and binder were fixed at 50 wt% and 5 wt%, respectively. Altering the magnesium/hexamine ratio had only little influence on the resulting color: Whereas formulation **1** was dominated by bright white light attributed to the highest magnesium percentage, all other formulations within this row showed a red-violet color with only little blue light emission in the outer flame parts (see Figure 1). Most of the formulations **1**–**10** achieved no constant burning after ignition and had to be re-ignited a few times.

Table 1. Indium-based formulations (in wt%) applying guanidine nitrate.

No.	GN [a]	In [b]	Mg [c]	Hexamine	Binder [d]
1	50	5	35	5	5
2	50	5	30	10	5
3	50	5	25	15	5
4	50	5	20	20	5
5	50	5	15	25	5
6	50	5	10	30	5
7	50	10	30	5	5
8	50	10	25	10	5
9	50	10	20	15	5
10	50	10	15	20	5

a) Guanidine nitrate. b) Indium (–100 mesh). c) Magnesium (50/100 mesh). d) Epoxy binder system (Epon 813/Versamid 140, ratio 4:1).

Figure 1. Burning of formulation **4**.

Potassium nitrate is a long-known, widely used component in pyrotechnics due to its availability in high purity and low costs, even though it suffers from hygroscopicity.[2b,3b] In contrast to guanidine nitrate, potassium nitrate decomposes at higher temperatures (T_{dec} = 400–750 °C).[27] Since the decomposition reaction is endothermic, the addition of metal fuels is mandatory to release enough energy for color emission.[2b,29] Applying potassium nitrate as oxidizer in formulations **11**–**20** resulted in violet burning formulations (Table 2, Figure 3). No

Table 2. Indium-based formulations (in wt%) applying potassium nitrate.

No.	KNO_3	In [a]	Mg [b]	Hexamine	Binder [c]
11	50	5	35	5	5
12	50	5	30	10	5
13	50	5	25	15	5
14	50	5	20	20	5
15	50	5	15	25	5
16	50	5	10	30	5
17	50	10	30	5	5
18	50	10	25	10	5
19	50	10	20	15	5
20	50	10	15	20	5

a) Indium (–100 mesh). b) Magnesium (50/100 mesh). c) Epoxy binder system (Epon 813/Versamid 140, ratio 4:1).

blue light was observed. Here again, formulation **11** had the shortest burning time (<1 s), which was due to the highest magnesium content.

The frequency measurement of formulation **15** proofed a non-regular burning behavior, which was detected by visual examination earlier (Figure 2 and Figure 3). After the first ignition and several flash reactions (= emission of light), the burning stopped and the pellet had to be re-ignited after a few seconds. This lack of the flash regularity as well as the insufficient flash separation, does not allow any potential application efforts. Another literature-known magenta strobe composition relies on cheaper compounds such as ammonium perchlorate, strontium nitrate, tetramethylammonium nitrate, hexamine and copper-aluminum alloy (50:50).[30] However, next to the application of cheaper but hazardous materials such as perchlorate, the color of this formulation was reported to substantially degraded in conditions of high humidity.

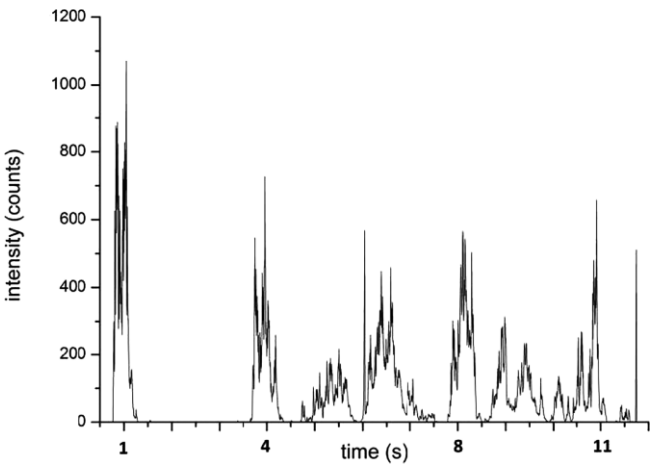

Figure 2. Frequency measurement of formulation **15** with 1 ms per scan.

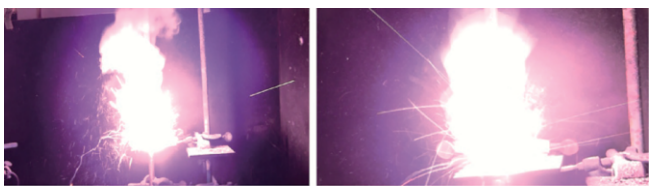

Figure 3. Burning of flare formulation **19** (left) and strobe formulation **15** (right).

Since the overall indium amount in formulations **11**–**20** (Table 2) was quite small compared to other colorants in existing formulations (typically more than 20 wt% up to 50 wt%), the effect of indium amounts up to 30 wt% was investigated next. Higher amounts of indium would result in even more expensive formulations, however, no blue light was observed for lower amounts. Formulations **21**–**29** applied 5-amino-1*H*-tetrazole (5-AT) instead of hexamine and potassium nitrate as an oxidizer (Table 3). This time, the effect of different oxidizer to metal fuel ratios was studied. Both 5-AT and hexamine were reported to give high spectral purities in light-producing pyrotechnical compositions before.[28] Magnesium is usually applied to increase the resulting burning temperature thus producing a higher incandescence level. Incandescence describes the phenomena of an emitted continuous spectrum covering the whole part of the visible spectrum (= white light), which can be observed for example with glowing steal ingots.[2a] This gain in luminous intensity comes along with a loss in spectral purity as the white light diminishes the color quality. To study the effect of lower magnesium content and reduced incandescence, formulation **24** was developed as a completely magnesium-free formulation. All of the tested formulations **21**–**29** revealed constant burning with a violet flame.

Table 3. Indium-based formulations (in wt%) applying 5-amino-1*H*-tetrazole.

No.	KNO_3	In [a]	Mg [b]	5-AT	Binder [c]
21	60	20	10	5	5
22	55	25	10	5	5
23	50	30	10	5	5
24	70	20	–	5	5
25	60	25	5	5	5
26	55	30	5	5	5
27	50	20	20	5	5
28	50	25	15	5	5
29	50	30	10	5	5

a) Indium (–100 mesh). b) Magnesium (50/100 mesh). c) Epoxy binder system (Epon 813/Versamid 140, ratio 4:1).

Reich and *Richter* did not provide any comparable wavelength values; instead they referred to an older dimensionless scale. In this scale, Na was set to the value 50, Sr to 104 and In_α/In_β to 110/147.[19] To obtain standardized values, the emission of selected formulations was measured with a calibrated spectrometer. A look at the emission spectrum of formulation **4** revealed five sharp lines and one broad signal in the range of 364–400 nm (UV/Vis region, violet) (Figure 4). Next to this, two sharp emission lines in the violet (410 nm) and the blue region (451 nm) were detected and referred to the In_α/In_β band as described by Reich and Richter earlier.[19]

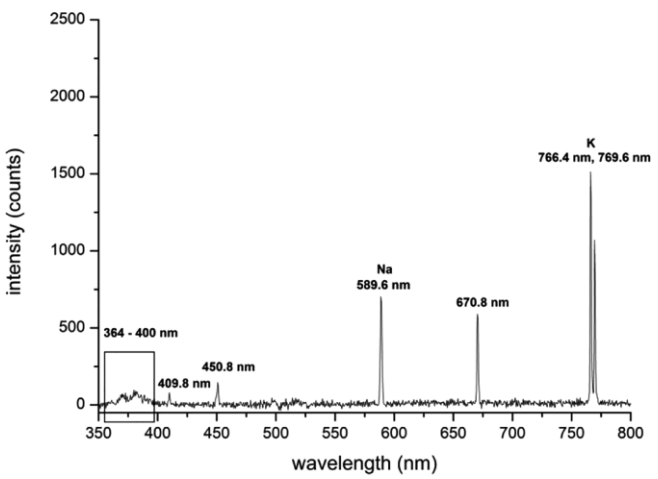

Figure 4. Emission spectrum of formulation **4**.

The remaining signals at 590 nm and 766 nm were assigned as sodium and potassium impurities. The last remaining line (671 nm) would fit as the main lithium emission. However, the

second emission of atomic lithium at 610 nm is missing and therefore, was ruled out. Due to the low intensity of the blue emission and the presence of the violet emission, it was concluded that it is not possible to achieve deep blue light with halogen-free indium-based formulations, since the impurities dominate the flame color.

A look at the emission spectrum of formulation **19** revealed higher intensities across the whole displayed wavelength range (Figure 5). The potassium line width increased and dominated the spectrum. This observation can be explained by a higher potassium concentration present in the flame, which is also known for other alkaline metals like lithium.[31] The broad emission at 496 nm was assigned to condensed MgO (s) particles.[32] Next to the identified emission lines of indium, sodium and potassium, several new unidentified lines at 404, 510, 517, 534, 581, 669, and 693 nm appeared. Again, the intensity of the emitted light in the blue region was too low to produce a blue color impression.

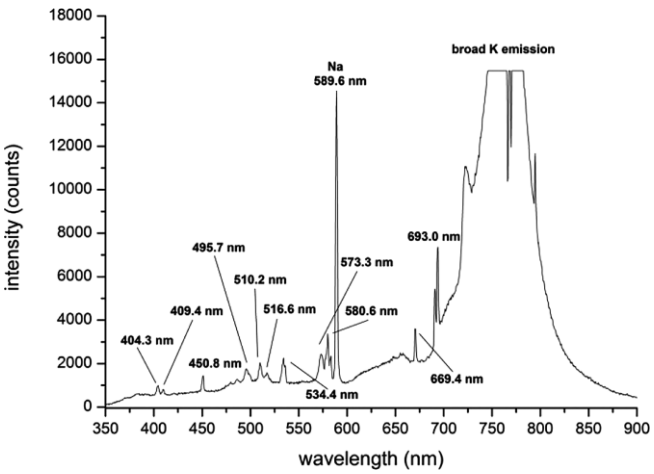

Figure 5. Emission spectrum of formulation **19**.

Finally, the color points of formulations **4**, **15**, and **19** are depicted in a CIE 1931 chromaticity diagram (Figure 6). As discussed before, the corresponding color impressions are in the violet region due to the high intensity of impurities.

Conclusions

Besides the well-known drawbacks of indium such as price and availability, we decided to investigate the flame chemistry from an academically point of view. For this reason, indium was introduced as colorant to a typical pyrotechnical formulation consisting of magnesium, hexamine or 5-amino-1*H*-tetrazole, potassium nitrate, or guanidine nitrate and an epoxy binder system. Unfortunately, none of the tested mixtures provided sufficient blue color emission. The main emission was dominated by magenta and violet, whereas only at the outer flame a blue color could be detected. Higher magnesium contents had higher luminous intensity, but at the same time a loss in spectral purity as a consequence. Due to the lack of existing flame emission spectra of indium, the author's measured emission spectra of selected indium-based pyrotechnical formulations. As a result, sharp lines at 410 nm and 451 nm could be detected, which were further referred to the In_α/In_β bands. However, the emission spectrum as well as the resulting flame color is dominated by impurities of sodium and potassium. In addition, the intensity of blue emission is insufficient and does not fulfill the quality demands for pyrotechnical formulations. Finally, it was concluded that it is not possible to observe a blue light with the provided indium-based halogen-free pyrotechnical system.

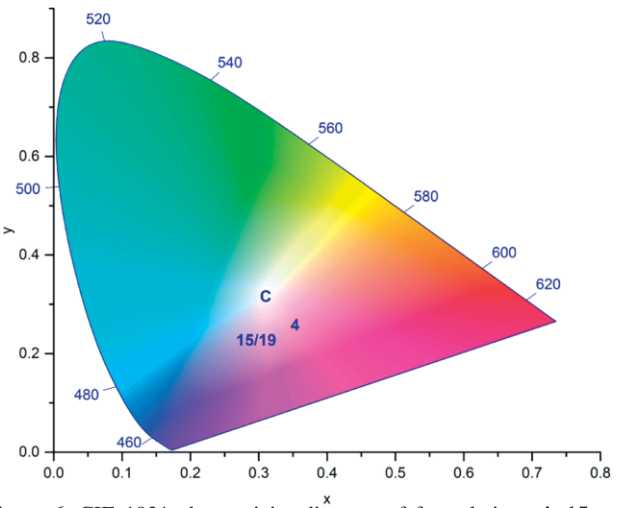

Figure 6. CIE 1931 chromaticity diagram of formulations **4**, **15** and **19**.

Experimental Section

CAUTION! The mixtures described here are potential explosives which are sensitive to environmental stimuli such as impact, friction, heat, and electrostatic discharge. While we encountered no problems in the handling of these materials, appropriate precautions and proper protective measures (safety glasses, face shields, leather coats, Kevlar® gloves, and ear protectors) should be taken when preparing and manipulating these materials.

Chemicals: The following materials were used as provided without further purification: Guanidine nitrate (98 %), Sigma–Aldrich; Potassium nitrate (99 %), Grüssing GmbH; Indium (99.9 %, –100 mesh), abcr Chemicals; Magnesium (99 %, 50/100 mesh), Grüssing GmbH; 5-Amino-1*H*-tetrazole (98 %), abcr Chemicals; Hexamethylenetetramine (99.5 %), abcr Chemicals; Epon 813, Hexion; Versamid 140, BASF.

Sample Preparation: All pyrotechnic samples were prepared in 1.0 g scale using the same procedure in order to ensure the reproducibility. Therefore, the different ingredients were weighed into a sample glass according to their respective weight percentages in the formulations. Each sample was transferred into a porcelain mortar and carefully ground to a homogeneous powder. After grinding, the binder solutions were added followed by a curing step. The so-prepared compositions were ground again and then, pressed into a cylindrical shape with the aid of a tooling die using a hydraulic press with a dead load of 2.0 t for 3.0 s.

Optical Measurement: Optical emissive properties were characterized using both an OceanOptics HR2000+ES spectrometer with an ILX511B linear silicon CCD-array detector (range 190–1100 nm) and

included software from OceanOptics. Spectra were recorded with a detector-sample distance of 1 m and an acquisition time of 1 ms per scan. The dominant wavelength (DW) and spectral purity (SP) were measured based on the 1931 CIE method using illuminant C as the white reference point. Five samples were measured for each formulation and all given values are averaged based on the full burn of the mixture. The controlled burn was filmed with a digital video camera recorder (SONY, DCR-HC37E).

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Keywords: Blue emitter; Indium; Emission spectrum; Pyrotechnic

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